

WE CLAIM:

1. A protective barrier coating for a silicon based substrate, comprising:
  - a diffusion barrier coating formed on a substrate, said diffusion barrier coating inhibiting or preventing diffusion of cations from the substrate to 5 an oxidation coating;
  - an oxidation barrier coating formed on the diffusion barrier coating, said oxidation barrier coating inhibiting or preventing the oxidation of the substrate; and
  - an environmental barrier coating formed on the oxidation barrier 10 coating, said environmental barrier coating inhibiting or preventing water vapor from reacting with the oxidation barrier coating to form volatile Si(OH)<sub>4</sub> and;
  - a thermal barrier coating formed on the environmental barrier coating, said thermal barrier coating limiting heat transfer from an environment to said environmental barrier coating and shielding the environmental barrier 15 coating from erosion and corrosive dust.
2. The protective barrier coating of claim 1, wherein said diffusion barrier coating is made of an over 99 mol % pure compound selected from the group consisting of SiC, Si<sub>3</sub>N<sub>4</sub>, and Si<sub>2</sub>ON<sub>2</sub>
3. The protective barrier coating of claim 1, wherein said oxidation barrier coating is made of a disilicate selected from the group consisting of Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, and Yb<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>.
4. The protective barrier coating of claim 1, wherein said oxidation barrier coating is made of a inner layer of Si<sub>2</sub>ON<sub>2</sub> and an outer layer of disilicate selected from the group consisting of Sc<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, and Yb<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>.

5. The protective barrier coating of claim 1, wherein the diffusion barrier layer is of  $\text{SiO}_2$  and the oxidation barrier layer is of disilicate selected from the group consisting of  $\text{Sc}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{Si}_2\text{O}_7$ , and  $\text{Yb}_2\text{Si}_2\text{O}_7$ .

6. The protective barrier coating of claim 1, wherein said environmental barrier coating includes a metallic disilicate and up to 10 weight % of a metallic oxide and monosilicate wherein the metal is one selected from the group of scandium, yttrium and ytterbium.

7. The protective barrier coating of claim 1, wherein said environmental barrier coating includes tantalum oxide alloyed with one oxide selected from the group of lanthanum oxide and alumina.

8. The protective barrier coating of claim 1, wherein said thermal barrier coating includes zirconia oxide alloyed with yttrium oxide.

9. The protective barrier coating of claim 1, wherein at least one interface between said diffusion barrier coating, oxidation barrier coating, environmental barrier coating and thermal barrier coating is graded to allow a gradual transition.

10. A protective barrier coating for a silicon based substrate, comprising;

an oxidation barrier coating formed on the substrate, said oxidation barrier coating comprising a metallic disilicate;

5 an environmental barrier coating formed on the oxidation barrier coating, said environmental barrier coating comprising a compound selected from the group consisting of a tantalum oxide alloy and a scandium silicate mixture.

11. The barrier coating of claim 10, wherein said metallic disilicate is at least one silicate selected from the group consisting of scandium, yttrium and ytterbia.

12. The barrier coating of claim 11, wherein said environmental barrier coating includes a metallic disilicate and up to 10 weight % of a metallic oxide or monosilicate of the same metal as the disilicate selected from the group of scandium, yttrium and ytterbium.

13. The barrier coating of claim 10, wherein the oxidation barrier layer has a duplex structure consisting of an inner layer of  $\text{Si}_2\text{ON}_2$  disposed between the silicon nitride substrate and the outer metallic disilicate layer.

14. The barrier coating of claim 10, wherein said tantalum oxide alloy is alloyed with an oxide, wherein the oxide is selected from one of the group consisting of about 4 to 7 mol% lanthanum oxide and about 1-3 mol % alumina.

15. The barrier coating of claim 10, wherein said metallic silicate is deposited on said substrate by at least one of the steps selected from the group consisting of spraying and dipping the substrate in a water based slurry of at least one of the disilicates selected from the group consisting of scandium, yttrium and ytterbium.

16. The barrier coating of claim 10, wherein the protective barrier coating includes a thermal barrier coating formed on the environmental coating, said thermal coating comprising stabilized zirconia and wherein said stabilized zirconia is yttria stabilized zirconia.

17. A combination silicon based substrate and a barrier coating comprising:

the silicon based substrate being selected from the group consisting of silicon carbide and silicon nitride,

5 a diffusion barrier coating in the range of 99 to 100% pure  $\text{Si}_3\text{N}_4$ ,  $\text{SiC}$  or  $\text{Si}_2\text{ON}_2$  on said substrate;

an oxidation barrier coating formed on the diffusion coating, said oxidation coating comprising a metallic disilicate;

10 an environmental barrier coating formed on the oxidation coating, said environmental coating made of a compound selected from the group consisting of a tantalum oxide alloy and scandium disilicate .

18. The combination of claim 17, wherein the oxidation barrier layer has a duplex structure consisting of an inner layer of  $\text{Si}_2\text{ON}_2$  disposed between the substrate and an outer metallic disilicate layer, the maximum thickness of the  $\text{Si}_2\text{ON}_2$  layer is not greater than 40 microns.

19. The combination of claim 17, wherein said metallic disilicate is at least one silicate selected from the group consisting of scandium, yttrium and ytterbia and said metallic silicate can contain up to 10 weight % of a metal oxide wherein said metallic silicate and said metallic oxide have the same base metal.

20. The combination of claim 17, wherein said tantalum oxide alloy is alloyed with an oxide wherein the oxide is selected from the group consisting of lanthanum oxide and alumina.

21. The combination of claim 17, wherein the protective barrier coating includes a thermal barrier coating formed on the environmental coating, said thermal barrier coating comprising stabilized zirconia.

22. A protective barrier coating for a silicon based substrate comprising;
  - a diffusion barrier coating formed on the substrate;
  - an oxidation barrier coating of scandium disilicate formed on the diffusion barrier coating and;
  - an environmental barrier coating formed on the oxidation barrier coating and comprising one of a tantalum oxide alloy and a mixture of scandium disilicate, scandium monosilicate and scandium oxide and;
  - the protective barrier coating also comprises a thermal barrier coating formed on the environmental barrier coating and comprising stabilized zirconia.
23. The protective barrier coating of claim 22, wherein said diffusion coating is in a range of about 0.5 to 10 micron thick layer of over 99% pure silicon based material selected from the group consisting of  $\text{Si}_3\text{N}_4$ ,  $\text{SiC}$  or  $\text{Si}_2\text{ON}_2$ .
24. The protective barrier of claim 22, wherein said oxidation barrier coating has a duplex structure consisting of an inner layer of  $\text{Si}_2\text{ON}_2$  disposed between the substrate and the outer metallic disilicate layer.
25. The protective barrier of claim 22, wherein said oxidation barrier coating is in the range of not greater than 40 microns thick.
26. The protective barrier of claim 22, wherein said oxidation barrier coating is in the range of not greater than 20 microns thick.
27. The protective barrier of claim 22, wherein said tantalum oxide alloy includes a metallic oxide selected from the group consisting of lanthanum oxide in the range of 3 to 10 mol % and alumina in the range of 1-3 mol %.

28. The protective barrier of claim 22, wherein said mixture of scandium disilicate, scandium monosilicate and scandium oxide includes scandium disilicate in a range of 60 to 100 weight percent.
29. The protective barrier of claim 22, wherein said environmental barrier coating is in a range of 5 to 50 microns thick.
30. The protective barrier of claim 22, wherein said thermal barrier coating includes stabilized zirconia in a range of 10 to 2000 microns thick.
31. The protective barrier of claim 28, wherein said thermal barrier coating includes stabilized zirconia in a range of 10 to 40 microns thick.
32. The protective barrier of claim 22, wherein said diffusion barrier coating is deposited by physical vapor deposition.
33. The protective barrier of claim 22, wherein said diffusion barrier coating is deposited by chemical vapor deposition.
34. The protective barrier of claim 22, wherein said diffusion barrier coating consists of one of the silicon compounds selected from the group consisting of SiC or Si<sub>3</sub>N<sub>4</sub>, in the range of 99 to 100% pure.
35. The protective barrier of claim 22, wherein said diffusion barrier coating, said oxidation barrier coating, said environmental barrier coating and said thermal barrier coating have a combined thickness in the range of 30-100 microns.

36. A method for protecting silicon based substrates comprising the steps of;
  - forming an oxidation barrier coating of metallic silicate on said substrate;
  - 5 forming an alloyed tantalum oxide environmental barrier coating over said oxidation barrier coating; and
  - forming a stabilized zirconia thermal barrier coating over said environmental barrier coating.
37. The method of claim 36, wherein the step of forming an oxidation barrier coating includes one of the steps selected from the group consisting of spraying or dipping said substrate in a water based slurry of metallic silicate to form a coat of metallic silicate on the substrate and then drying and sintering 5 said coat of metallic silicate.
38. The method of claim 37, wherein the step of sintering includes heating in a vacuum to a temperature of about 1000°C (1800°F), switching to an inert atmosphere, and sintering the substrate and the coat of metallic silicate at a temperature in a range of about 1400-1650°C (2550-3000°F).
39. The method of claim 36, wherein the step of forming an oxidation barrier coating includes a step of forming a pre-oxidized surface on the substrate by heating the substrate in air at about 1200 to 1400°C (2200-2550°F).
40. The method of claim 39, wherein the step of forming an oxidation barrier coating includes a step of coating said pre-oxidized surface with metallic silicate powder to form a powder coating and then sintering said powder coating.

41. The method of claim 36, wherein the step of forming an oxidation barrier coating includes a step of oxidizing a surface of said substrate to form a layer of silica on said substrate, dipping the substrate in a sol gel solution to form a film on said substrate, withdrawing the substrate from the sol gel solution and placing it in moist air where metallic hydroxides in said film react with the moist air to form a metallic oxide coat which is then sintered.

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42. The method of claim 41, wherein the step of forming an oxidation coating includes drying said coat of metallic oxide at about 200-600°C (390-1100°F) and then sintering said coat at a temperature in the range of about 600-1650°C (1100- 3000°F) to form a metallic silicate on said substrate.

43. The method of claim 42, wherein the step of sintering includes crystallizing a silicate grain boundary in said substrate.

44. The method of claim 36, wherein the metallic silicate is one silicate selected from the group consisting of  $\text{Sc}_2\text{Si}_2\text{O}_7$ ,  $\text{Sc}_2\text{SiO}_5$ ,  $\text{Y}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{SiO}_5$ ,  $\text{Yb}_2\text{Si}_2\text{O}_7$  and  $\text{Yb}_2\text{SiO}_5$ .

45. A method for protecting silicon based substrates comprising the steps of:

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forming an oxidation barrier coating on said substrate, wherein the step of forming the oxidation barrier includes the step of sintering said oxidation barrier and substrate in a wet gas containing hydrogen.

46. The method of claim 45, wherein the step of forming an oxidation barrier coating includes dipping said substrate in a water based slurry of metallic silicate to form a coat of metallic silicate on the substrate or of spraying said

water based slurry on the substrate then drying and then sintering said coat of  
5 metallic silicate to a temperature in a range of about 1250-1600°C (2280-  
2910°F).

47. The method of claim 45, wherein the step of forming an oxidation barrier coating includes dipping said substrate in a water based slurry of metallic silicate to form a coat of metallic silicate on the substrate or of spraying said water based slurry on the substrate then drying and then sintering said coat of  
5 metallic silicate to a temperature in a range of about 1300-1550°C (2370-  
2820°F) in a flowing gas made up of 0 to 10% hydrogen in an inert gas.

48. The method of claim 45, wherein the step of forming an oxidation barrier coating includes dipping said substrate in a water based slurry of metallic silicate to form a coat of metallic silicate on the substrate or of spraying said water based slurry on the substrate then drying and then sintering said coat of  
5 metallic silicate to a temperature in a range of about 1300-1550°C (2370-  
2820°F) in a flowing moist gas of made up of 0 to 10% hydrogen in an inert gas.

49. A method for protecting turbine engine components made from silicon based substrates comprising steps of;  
                forming a diffusion barrier coating on said substrate;  
                forming an oxidation barrier coating of metallic silicate on said  
5 diffusion barrier coating;  
                forming an environmental barrier coating over said oxidation barrier coating;  
                forming a stabilized zirconia thermal barrier coating over said environmental barrier coating; and  
10                 wherein said metallic silicate is from the group of silicates

$\text{Sc}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{SiO}_5$  and  $\text{Yb}_2\text{Si}_2\text{O}_7$ .

50. The method of claim 49, wherein the step of forming an oxidation barrier coating includes applying a water based slurry of metallic silicate to said substrate to form a coating which is sintered to form a layer of metallic silicate on the substrate.

51. The method of claim 49 wherein the step of forming an oxidation barrier coating includes a step of pre-oxidizing the substrate by heating the substrate in air.

52. The method of claim 49, wherein the step of forming an oxidation barrier coating includes a step of oxidizing said substrate to form a layer of silica on said substrate, dipping the substrate in a sol gel solution to form a film on said substrate, withdrawing the substrate from the solution and placing it in moist air where metallic hydroxides in said film react with the moist air to form a metallic oxide coat on said substrate.

53. The method of claim 52, wherein the step of forming an oxidation barrier coating includes drying said coat of metallic oxide and then sintering said coat of metallic oxide at a temperature less than a melting point of the coat of metallic oxide to form a metallic silicate on said substrate.

54. The method of claim 49, wherein the step of forming an oxidation barrier coating includes deposition of an duplex coating composed of an inner  $\text{Si}_2\text{ON}_2$  layer and an outer layer metallic silicate on said diffusion barrier coating.

55. The method of claim 49, wherein the step of forming an oxidation

barrier coating includes deposition of an duplex coating composed of an inner Si<sub>2</sub>ON<sub>2</sub> layer and an outer layer metallic silicate on said diffusion barrier coating and of sintering both the inner and outer layer in a furnace with a wet hydrogen atmosphere in a temperature range from 1250 to 1600°C (2280-2910°F).  
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56. The method of claim 49, wherein the step of forming the inner Si<sub>2</sub>ON<sub>2</sub> layer and an outer layer metallic silicate includes plasma spraying.

57. The method of claim 49, wherein the step of forming the inner Si<sub>2</sub>ON<sub>2</sub> layer and an outer layer of metallic silicate includes electron beam-physical vapor deposition.

58. The method of claim 49, wherein the step of forming an environmental barrier coating includes plasma spraying.

59. The method of claim 49, wherein the step of forming an environmental barrier coating includes electron beam-physical vapor deposition.

60. A method for protecting silicon carbide substrate comprising the steps of:

forming a diffusion barrier coating on said silicon carbide substrate and;

5 forming an oxidation barrier coating of metallic silicate on said diffusion barrier coating;

forming an environmental barrier coating over said oxidation barrier coating;

10 forming a stabilized zirconia thermal barrier coating over said environmental barrier coating.

61. The method of claim 60, wherein the metallic silicate is a rare earth silicate and the rare earth silicate is from the group of silicates consisting of ytterbium and yttrium.

62. The method of claim 60, wherein the step of forming an oxidation barrier coating includes the step selected from the group consisting of spraying and dipping said substrate in a slurry of metallic silicate to form silicate on the diffusion coating.

63. The method of claim 62, wherein the step of forming a diffusion barrier coating includes depositing a coat of one compound selected from the group consisting of SiC, Si<sub>3</sub>N<sub>4</sub>, or Si<sub>2</sub>ON<sub>2</sub> on said substrate.

64. The method of claim 61 wherein the step of forming said oxidation barrier coating can include joining said substrate to a second substrate.

65. A method for protecting silicon nitride component substrates comprising the steps of:

- forming a diffusion barrier coating on said substrate;
- forming an oxidation barrier coating of scandium silicate on said 5 silicon nitride substrate;
- forming an alloyed tantalum oxide environmental barrier coating over said oxidation coating.

66. The method of claim 65 further comprising the step of:  
forming a stabilized zirconia thermal barrier coating over said environmental coating.

67. A silicate based composition for joining together at least two ceramic components, said composition comprising: a metallic silicate from the

group of silicates including;  $\text{Sc}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{Si}_2\text{O}_7$ ,  $\text{Y}_2\text{SiO}_5$  and  $\text{Yb}_2\text{Si}_2\text{O}_7$ .

68 The silicate based composition of claim 67 wherein said silicate composition is formed by reacting constituents of a mixture applied to a surface of a substrate, and wherein said silicon-based composition is bonded to a substrate surface of each said ceramic component.

69. The silicate based composition of claim 68 wherein said silicate based composition further comprises a dispersion of particles selected from the group of silicon nitride and silicon oxynitride.

70. The silicate based composition of claim 68, wherein said surface of said substrate comprises a surface of a first ceramic component, said silicate based composition is used as a braze, for joining said first ceramic component to a second ceramic component.

71. The silicate based composition of claim 70, wherein at least one of said first and second ceramic components comprises a silicon based component, and wherein said first and second ceramic components are adapted for use at elevated temperatures up to about 2400° F (1315°C).